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Electron-phonon interactions in energetic displacement cascades

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The role of electron-phonon interactions in energetic displacement cascades in metals is evaluated by considering the diffusion of electrons in a localized heat spike. It is found that the electronic system in this region always falls out of equilibrium with the lattice at long times and that whether electronic equilibrium is achieved at short times depends almost catastrophically on the parameters that describe the electron-phonon interaction. Neglect of the electron-phonon coupling may lead to overestimates of thermal spike temperatures in many metals.

Theoretical descriptions of radiation damage in solids, wherein fast particles leave behind trails of displaced atoms, have focused almost exclusively on energetic collisions between the incident particle and the massive atomic cores of the target solid. The role of the valence electrons has largely been ignored, other than as an energy sink that contributes to the ability of the material to slow down the primary particle and, to a much smaller extent, to damp the motion of secondary recoil atoms. A key factor in considering the role electron-phonon coupling is the recent evidence that thermal spikes are an important ingredient in the damage process.[1,2] In the thermal spike model the energy deposited in a locality by a collision is quickly partitioned among neighboring atoms, raising the region to a very high temperature; the radius of the thermal spike subsequently increases and the temperature decreases as the heat spreads by thermal conduction.[3] That very high temperatures are achieved is illustrated by the example of a 10-keV collision which leaves 1 eV/atom (10^4 K) even when spread over 10^4 atoms ($r \approx 30$ Å). Still higher energy densities are realized at earlier times and in materials with high atomic number at higher recoil energies. In Ag, for example, the energy density associated with a 10-keV recoil is ≈ 10 eV/atom after 0.1 ps (Ref. 4). Heat loss from this excited region brings the temperature below the melting point of the material only after several ps (Ref. 1). We demonstrate here that these times are comparable to those for electronic thermalization and that neglect of the heat capacity and thermal conductivity of the electronic system can lead to incorrect evaluations of thermal spike in many metals.

Consider first the way a spike of hot lattice, radius r, temperature T, total energy Q, can heat up the local electronic system. The unexpected results that we shall describe are as follows: The electronic system is always out of equilibrium with the lattice at very long times, and the question of whether or not the electronic equilibrium is achieved at short times, with r small, depends with almost catastrophic sensitivity on the parameters that describe the electron-lattice interaction. The first of these points is readily made. For definiteness, we suppose we are concerned with a metal of Wigner-Seitz radius r_s for which the electronic mean free path depends linearly on T as predicted by the harmonic approximation. The result may be written

$$\lambda = r_s T_0 / T \quad (T > \Theta_D), \tag{1}$$

in which T_0 is the value of T at which λ is reduced to r_s, and Θ_D is the Debye temperature. Energy conservation among the $N = (r/r_s)^3$ hot atoms yields, for a classical system,

$$r = r_s (Q/3k_B T)^{1/3}. \tag{2}$$

Its stronger T dependence makes $\lambda > r$ for some low T. Therefore, at sufficiently long times, the electronic system cannot equilibrate with the lattice. For $Q \approx 10^4$ eV and $T_0 \approx 10^5$ K (Cu), $\lambda \approx r$ for $T \approx 5000$ K, where the classical approximation used here cannot be questioned. Thus, for this example, the local electronic system remains much cooler than the lattice during most of the equilibration period.

Whether or not equilibrium is achieved at *short* times depends very sensitively on the electron-phonon coupling measured by T_0. This happens because as λ is reduced it becomes progressively more difficult for carriers to "random walk" out of the hot volume of radius r. The number of free paths required to escape the cascade zone is clearly $\approx (r/\lambda)^2$; and if the carriers absorb an energy $k\Theta_D$ per

collision,[5] their effective local temperature reaches

$$T_{\text{eff}} = \Theta_D r^2/\lambda^2 . \quad (3)$$

Equations (1)–(3) can be solved for $T_{\text{eff}} = T$ to find that electronic equilibrium is achieved for radii smaller than a critical value

$$r_c = r_s \Theta_D Q/3k_B T_0^2 , \quad (4)$$

when the temperature has cooled to

$$T_c = 9T_0^6/\Theta_D^3 (Q/k_B)^2 . \quad (5)$$

Atoms of moderate atomic weight, like Cu and Ni, have thermal spikes with maximum energy of ≈ 10–20 keV due to subcascade formation at higher energies.[6] For materials with heavier nuclei, the energy may be much larger. The resistivities of Cu and Ni indicate values of 4.5×10^4 and 1.5×10^4 K, respectively, for T_0. Equation (5) with $\Theta_D \approx 300$ K then predicts critical temperatures of 2×10^5 K for Cu and 300 K for Ni. We conclude that materials as similar as Cu and Ni may behave very differently from each other during thermal spikes. In particular, Ni achieves electronic equilibrium essentially throughout the entire duration of the spike whereas in Cu the electronic system may never equilibrate with the lattice. Note that the preceding results are not expected to be accurate above temperatures T_0 at which the mean free path is reduced to atomic dimensions. However, this limitation does not affect the conclusion just stated. The importance of the results lies in the fact that the electronic specific heat of Ni equals the lattice specific heat at $T \approx 10^4$ K, and at 10^5 K may be a factor of 10 larger, when the $4s$ and nine $3d$ electrons are all excited. Electronic excitation thus serves to moderate the temperature achieved in the hot early stages of the spike and to increase the heat conduction out of this zone.

The predictions of the simple equations above require some modification in real metals at the extreme temperatures of the thermal spike. For Cu and Ni, in particular, the electronic structures are very similar when blurred over an energy scale of several eV. One may therefore expect that properties such as the electronic heat capacity and electron scattering times of these metals become much less different at temperatures where the Fermi edge width exceeds 1 eV. This has been verified in explicit calculations by Levy, Barak, and Ashkenazi.[7] In the case of Ni, the reduced electronic specific heat at high temperatures will somewhat decrease the effectiveness of the electronic equilibration in moderating the thermal spike temperatures. Our schematic discussion indicates, however, that the electronic system of Cu cannot be heated through the low-temperature regime $< 10^3$ K, so that the changes of properties relevant to $T \approx 10^4$ K play no part in the thermal spike evolution of Cu. Thus, the qualitative distinction between Cu and Ni is maintained. In other cases, of course, the electronic properties may be less temperature dependent than Cu and Ni, for example, when the Fermi level lies in the middle of broad d bands.

One may also reasonably inquire whether or not heat conduction by quaisparticle transport is sufficiently rapid to carry off the electronic energy before the electronic system is fully heated. To answer this we note that the number of excitations per second in a volume v, temperature T_{eff}, must be $k_B T_{\text{eff}} N(E_F) v/\tau$, with $N(E_F)$ the electron density of states at the Fermi level and τ the scattering time for each of the $kT_{\text{eff}}N(E_F)v$ excitable states. With $k\Theta_D$ of energy per excitation and T_{eff} taken from Eq. (3), the power absorbed by the electrons from the lattice is

$$P \approx (k\Theta_D)^2 (r^2/\lambda^2) N(E_F) 4\pi r^3/3\tau . \quad (6)$$

Using Eqs. (1) and (2), this may be rewritten for $r = r_c$ in terms of the electronic thermal conductivity $\kappa(T)$ as

$$P = 4\pi r^2 \kappa(T) (dT/dr)_{\text{eff}} , \quad (7)$$

with

$$(dT/dr)_{\text{eff}} = (T/r_0)(\Theta_D^2 Q T/\pi^2 k_B T_0^4) . \quad (8)$$

The power dissipated by conduction electrons may thus keep pace with the power absorbed for reasonable values of an "effective temperature gradient" $(dT/dr)_{\text{eff}}$. On substituting values for Cu at $r = r_c$ and $T = 10^4$ K with $Q = 10$ keV, one finds $(dT/dr)_{\text{eff}} \approx 10^{-3} T/r_0$, whereas for Ni the value is $\approx T/10r_0$. Thus, the absorbed heat is readily dissipated for 50-Å Cu spikes but not for Ni, in which the required temperature gradient exceeds T/r, so that the electrons in the spike presumably heat into local equilibrium with the lattice as describe above.

The above analysis is based on the harmonic approximation for lattice vibrations; the linear extrapolation of the electronic mean free path above the melting temperature of the material, therefore, is not wholly justified. The above concepts are, nevertheless, valid, and to judge whether or not a particular metal should be affected by electron-phonon coupling requires only comparing the electron mean free path in the liquid metal to the dimensions of the cascade. Electron mean free paths have been calculated for the liquid metals, Cu, Ni, Co, and Fe with the results $\lambda = 21.6$, 8.1, 2.1, and 0.5 Å, respectively.[8] Although these values should be considered only as estimates of the true mean free paths, rather than observed values, they do substantiate the qualitative findings that electron mean free paths can be smaller than the cascade dimensions in many metals. We expect that electron-phonon coupling should play an important role in the cascade dynamics of such metals.

Rapid equilibration between electron and phonon systems should moderate the effects of thermal spikes since, as mentioned above, the maximum spike temperature can be drastically reduced by the heat capacity of the electron system and the heat loss by thermal conduction increased. Indeed, those metals which are expected to have strong coupling, i.e., those with high $N(E_F)$, do not show strong influences of thermal spikes in ion beam mixing[9] and cascade collapse[10] experiments. In the past, this has been attributed solely to the higher melting temperatures of these metals.[2,9] Attempts to observe heating of the electronic system in cascades more directly through measurements of thermionic emission from cascade zones have not proved successful.[11] Evaluation of these measurements, however, neglect the point established in this paper, that the heat capacity of the electronic system reduces the spike temperature and consequently the electron current.

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